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Total number of authors:
13

Published in:
Proceedings of SPIE

Link to article, DOI:
[10.1117/12.2592717](https://doi.org/10.1117/12.2592717)

Publication date:
2021

Document Version
Publisher's PDF, also known as Version of record

[Link back to DTU Orbit](#)

Citation (APA):

Wang, Y., Adamu, A. I., Habib, M. S., Dasa, M. K., Petersen, C. R., Antonio Lopez, J. E., Zhou, B., Jepsen, P. U., Schülzgen, A., Bache, M., Correa, R. A., Bang, O., & Markos, C. (2021). Ultraviolet to mid-infrared gas-filled anti-resonant hollow-core fiber lasers. In K. Kalli, A. Mendez, & P. Peterka (Eds.), *Proceedings of SPIE* (Vol. 11773). [1177308] SPIE - International Society for Optical Engineering. Proceedings of SPIE - The International Society for Optical Engineering Vol. 11773 <https://doi.org/10.1117/12.2592717>

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SPIE.

Event: SPIE Optics + Optoelectronics, 2021, Online Only

Ultraviolet to mid-infrared gas-filled anti-resonant hollow-core fiber lasers

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ABSTRACT

We will present our recent works on fiber lasers enabled by noble and Raman-active gas-filled anti-resonant hollow-core fiber (ARHCF) technology. First, we will present the generation of supercontinuum (SC) spanning from 200 nm to 4 μ m based on a Argon (Ar)-filled ARHCF pumped at 2.46 μ m wavelength with 100 fs pulses and ~ 8 μ J pulse energy. Then we will discuss our recent work on stimulated Raman scattering (SRS) effect in a hydrogen (H_2)-filled ARHCF, to achieve infrared Raman lasers. By employing the single-stage vibrational SRS effect, a 4.22 μ m Raman laser line is directly converted from a linearly polarized 1.53 μ m pump laser. A quantum efficiency as high as 74% was achieved, to yield 17.6 μ J pulse energy. The designed 4.22 μ m wavelength is overlapped with the strongest CO_2 absorption, therefore constituting a promising way for CO_2 detection. In addition, we report a multi-wavelength Raman laser based on cascaded rotational SRS effect. Four Raman lines at 1683 nm, 1868 nm, 2099 nm, and 2394 nm are generated, with pulse energies as high as 18.25 μ J, 14.4 μ J, 14.1 μ J, and 8.2 μ J, respectively. The energy of these Raman lines can be controlled by tuning the H_2 pressure from 1 bar to 20 bar.

Keywords: Anti-resonant hollow-core fiber, supercontinuum, Raman laser.

1. INTRODUCTION

The recent advent of silica ARHCF technology has enabled new research directions towards development of novel lasers from deep-ultraviolet (DUV) to the mid-infrared (MIR)¹. The laser beam is confined within the hollow-core region of ARHCF through anti-resonant effect, therefore possessing important advantages of gas medium such as low loss, broad transmission range, and high damage threshold. The easy control of gas pressure and species allows the flexible tailoring of fiber nonlinearity and dispersion properties. These factors make the ARHCF an ideal platform for the propagation of high peak power and energy laser pulse with rich nonlinear dynamics such as four-wave mixing², pulse compression³, and resonant dispersive wave (DW) emission⁴. An influential result of these nonlinear effects is the SC generation broadly spanning from DUV to MIR spectral region⁵. In 2019, we reported that pumping a 30 cm long Ar-filled ARHCF under 30 bar pressure with a 2460 nm laser with ~ 100 fs pulse duration and ~ 8 μ J pulse energy, to generate a SC from 200 nm up to 4 μ m⁶. Further investigations have been performed in recent to achieve a tunable DUV SC by pumping an Ar-filled ARHCF with a 1030 nm laser with a pulse energy of 24.2 μ J and compressed pulse duration of 30 fs⁷. As a result, the center wavelength of DW can be tuned from 236 to 377 nm by adjusting the Ar pressure from 2.2 to 5.7 bar.

SRS is another important nonlinear effect which occurs as a result of the intense interaction between laser pulse and a Raman active gas-filled in ARHCF^{8,9}. The SRS effect leads to the emission of Stokes and anti-Stokes waves, and are widely used for laser frequency down- and up-shifts. Phase matching condition of the anti-Stokes emission can be satisfied with the pressured dependent fiber dispersion property and the dispersion difference of different order modes^{10,11}. Contrary to SC generation, the gas-filled fiber Raman laser can achieve a narrow linewidth in few GHz level and even hundreds MHz level¹¹⁻¹³. This is because SRS process in gas has a narrow Raman gain bandwidth¹⁴, and can occur at a lower laser intensity compared to other nonlinear effects such as self-phase modulation (SPM). By employing vibrational SRS effect

in gases such as H_2 and methane (CH_4) with a long Stokes frequency shift coefficient¹⁵, MIR single-wavelength laser can be directly converted from the mature near-infrared (NIR) region. Several impressive reports on MIR fiber Raman lasers have already been reported, based on the vibrational SRS process in H_2 and CH_4 ^{16–19}. In these reports, the quantum efficiency can easily reach a desirable level (e.g., >60%), leading to high pulse energies in micro-joule range. Multi-wavelength (or frequency comb-like) Raman lasers due to cascaded (anti-) Stokes effects have also been reported^{9,18,20–22}, which have versatile applications such as sensing, wavelength division multiplexing communication, and optical signal processing²³. In comparison with the conventional method of selecting multiple wavelengths with a comb filter within the limited gain bandwidth of a rare-earth ion-doped gain medium²⁴, this method can achieve multi-wavelength operation in a much broader wavelength range.

In this paper, we present our recent works regarding gas-filled ARHCF lasers, including a SC generation from 200 nm to 4 μm , a MIR Raman laser generation at 4.22 μm with high energy of 17.6 μJ and 4.22 μm , and a multi-wavelength Raman laser spanning from 1.53 μm to 2.4 μm .

2. SC GENERATION BASED ON AR-FILLED ARHCF

Figure 1(a) shows the experimental setup of SC generation. A Ti:sapphire amplifier was used to pump an OPA with a few millijoule pulse energy to generate a MIR pump. The MIR pump is a linearly polarized 100 fs pulses at 1 kHz repetition rate with a central wavelength of 2460 nm and its power was controlled by neutral density (ND) filters and by rotating a nanoparticle linear film polarizer (P1). The pump laser is coupled into a 30 cm long ARHCF through a plano-convex lens with 5 cm focal length. Both ends of the ARHCF are sealed by two gas cells for Ar filling. Optical windows and lenses used in this setup were uncoated CaF_2 to obtain >90% transmission from 200–5000 nm. The laser at the output of the fiber is collimated and directed to either a CCD-based detector (Oceanoptics HR2000+) or to an infrared Spectrometer by using a flip-mirror, to measure the output spectrum at 183–1100 nm and 1000–5000 nm, respectively.

The ARHCF consists of seven non-touching silica capillaries with wall thickness of ~640 nm forming a core with diameter of ~44 μm , as shown in the scanning electron microscope (SEM) images of Figure 1(a). The propagation loss of the ARHCF is simulated based on a finite element method (FEM) using Comsol software with contributions from the material (silica), mode confinement, and surface scattering loss. According to the simulation result in Figure 1(b), one can see that the ARHCF transmits light over a broad spectral range with the anti-resonant window spanning from ~1400 nm to ~4000 nm, which is consistent with the measured loss spectrum based on cut-off method. Higher order anti-resonant windows allow for light to propagate even in the DUV range close to the bandgap of fused silica. The refractive index used in the calculations was based on the Sellmeier equation found in Ref. [25], which is valid from 210 nm up to 3.7 μm . However, it is important to mention that careful consideration of the silica refractive index is required for wavelengths less than 210 nm due to strong electronic bandgap absorption²⁶. To this end, the measured refractive index data of silica from Ref. [26] was used to calculate the GVD and confinement loss for wavelengths less than 210 nm. Figure 1(c) shows the calculated GVD of the fiber from 200 nm to 3700 nm for different gas pressures from 0 to 30 bar. When the pressure of the gas in the fiber increases it counterbalances the anomalous dispersion of the fiber, and as a result the ZDW shifts towards longer wavelengths and the nonlinear refractive index n_2 increases, as it has been extensively described elsewhere^{1,27,28}.

Before the start of the experiments, the fiber was purged several times with high purity Ar to remove any remaining atmospheric air and other impurities. Initial increase of the pump power is associated with the SPM dominated spectrum broadening, which is found to be weakly dependent on pressure. When the spectrum broadens to visible region, it becomes pressure sensitive due to the significant change of the GVD in the visible compared to the MIR. The pressure has a more pronounced effect on the MIR and UV-visible regime at higher pump pulse energy. At the maximum pulse energy of 20 μJ , the increase of Ar pressure first leads to significant broaden toward UV region with a dispersive wave at 240 nm. When the pressure is above 15 bar the long-wavelength edge starts to quickly increase, meanwhile the energy of the other DW around 275 nm slowly rises up to replace the DWs around 240 nm. The increasing MIR broadening and 275 nm DW generation is attributed to the increasing nonlinearity, while the diminishing 240 nm DW generation is believed to be caused by the red-shifting of the ZDW. Figure 1(d) shows the measured SC spectrum spanning from 200 nm up to 4 μm at the maximum pulse energy of 20 μJ . The main limitation of the spectral extension towards the MIR is the increasing propagation losses, reaching 100 dB/m at 3500 nm from which silica multiphonon absorption is the dominant loss mechanism. To improve the MIR transmission it is therefore paramount to reduce the mode-field overlap with the silica structure to a minimum, which can be achieved through fiber geometric modification.

Figure 1. SC generation based on Ar-filled ARHCF. (a) Experimental setup. Insets in bottom are SEM images of the ARHCF. The components of the setup are: neutral density filter (ND), protected silver mirrors (M1-2), linear film polarizer (P1), CaF_2 plano-convex lenses (L1-2), flip mirror (F1), and a gold coated parabolic mirror (PM1), 30 cm of ARHCF. (b) Simulated losses (blue) of the ARHCF, including contributions from surface scattering, mode confinement, and silica material loss. A measured loss spectrum based on cut-off method is provided for comparison. (c) Calculated group velocity dispersion (GVD) for the ARHCF at different levels of Ar pressure (solid lines). The measured dispersion of the fiber in the MIR is shown in circles for comparison (measured at ambient air condition). (d) Measured spectrum of SC obtained by pumping the ARHCF with 100 fs pulses at 2460 nm (anomalous dispersion region).

3. MIR RAMAN HIGH ENERGY RAMAN LASER GENERATION

Figure 2(a) shows the experimental setup of Raman laser generation. The pump laser is a master oscillator power amplifier (MOPA) based on a directly modulated distributed feedback diode laser. The seed oscillator delivers a linearly polarized pulse train at a repetition rate of 8 kHz with 6.9 ns pulse duration. The center pump wavelength was measured to be 1532.8 nm, corresponding to a Raman laser wavelength of 4.22 μm . The seed laser is first pre-amplified using two stages of core-pumped Er-doped fiber (EDF) pre-amplifiers sharing the same CW pump laser diode. The final amplification stage is a co-doped double-cladded Er/Yb fiber cladding-pumped by a 10 W CW laser diode at 915 nm. The large core size of 25 μm of the Er:Yb-doped fiber effectively suppresses the laser linewidth broadening from nonlinear effects. Band pass filters with ~ 2 nm bandwidth were used to suppress the ASE after each amplifier. After the power amplifier, the maximum average power was measured to be 1.41 W (including ASE). According to the spectral intensity distribution shown in Figure 2(a), the average power of the pulse signal is estimated to be 643 mW, corresponding to 11.6 kW peak power and 80.4 μJ pulse energy. The polarization extinction ratio (PER) of the pump laser is measured to be -19 dB, which includes the contributions of both the pulse signal and the ASE. Because the ASE generated during the amplification process is intrinsically un-polarized, the pulse signal in isolation is expected to have higher PER. The laser linewidth was measured by an optical spectrum analyzer (ANDO AQ6317B, AssetRelay) with a resolution of 0.01 nm to be 0.06 nm.

H_2 is filled into a piece of ARHCF using two gas cells. A half-wave plate (HWP) is placed before the ARHCF to optimize the laser polarization direction and maximize the Raman laser power²⁹. The output beam from the pump laser was collimated and coupled into the ARHCF through a pair of plano-convex lenses. The coupling efficiency was $\sim 81\%$. At the output side of the ARHCF, a 2.4 μm long-pass filter with 20% transmission loss at 4.22 μm is used to extract the vibrational Raman line laser from the residual pump. Figure 2(b) shows the SEM images of the ARHCF. The cladding consists of a ring of 7 capillaries with a wall thickness of 1030 nm and a diameter of 43.6 μm , forming an air-core region with a diameter of ~ 73 μm . This structure enables two transmission bands separated by a strong loss peak centered at 2.0 μm , as can be seen in the simulated loss spectrum depicted in Figure 2(c). The simulation was done based on the finite-element method using software COMSOL with parameter settings identical to those in Ref. [30], but without taking into

consideration the surface scattering loss due to the small surface roughness of the silica waveguide in the infrared region and the large core diameter of the fiber³¹. The measured loss at 1.53 μm is ~ 0.05 dB/m obtained via a cut-back method using the pump laser as light source. The fiber loss in the 2.5–4.5 μm region was also measured with a cut-back method, but using a broadband MIR SC source (MIR-SuperK, NKT Photonics). The loss at 4.22 μm could not be precisely obtained by the cut-back measurement because of the CO_2 presence in the ambient air inside the fiber, but from the surrounding background loss we can see it is approximately 1.5 dB/m. Note that the loss difference between the simulation and experiment slowly increases toward shorter wavelengths. This effect is due to the wavelength-dependent bend loss of the fiber (see Eq. (4) in Ref. [32], where the minimum bend radius is inversely proportional to the square of wavelength).

Figure 2. MIR Raman laser generation in H_2 -filled ARHCF. (a) Experimental setup. (b) SEM images of the ARHCF. (c) Simulated (brown curve) and measured (blue curve) loss spectra of the ARHCF. The red star indicates the measured loss at the pump wavelength of 1532.8 nm using the cutback method. (d) Spectra of pump and Raman laser. Top axis shows the absorbance spectrum of CO_2 obtained from the high-resolution transmission molecular absorption database (HITRAN). (e) Evolution of average Raman laser power and residual pump power as a function of coupled pump power. The dash line shows the quantum limit of the Raman laser. (f) Calculated quantum efficiency and pulse energy corresponding to (e).

The length of the ARHCF was optimized to 3.95 m, to maximize the Raman laser power at the maximum pump power. In this condition, it is found that the Raman power scales up with increasing H_2 pressure and steadily reaches saturation power at ~ 141 mW when the pressure approaches 20 bar. This trend can be attributed to the known saturation of the Raman gain at high pressure and decreased dephasing time of H_2 ³³. Figure 2(d) depicts the spectrum at 20 bar measured using an optical spectrometer (Spectro 320, Instrument Systems) with a resolution of 0.05 nm, where a strong laser line at 4.22 μm is formed as a result of the vibrational Raman Stokes shift from the pump laser. A rotational Raman line is also formed at 1.68 μm , but its intensity is negligible when compared to the vibrational Raman line as a result of the optimized pump polarization³⁴. Simulated absorbance spectrum of CO_2 is also provided for comparison in this figure. It can be seen that the spectrum of Raman laser is well located in the R-branch of absorption spectrum of CO_2 molecules, suggesting the great usability of this laser for high sensitivity CO_2 monitoring. The dependences of the Raman laser power and residual pump power on the coupled pump power are measured and shown in Figure 2(e). By increasing the coupled pump power, the power of the Raman laser continuously increases while its growth rate gradually decreases to a nearly constant value. Correspondingly, the calculated quantum efficiency in Figure 2(f) becomes nearly independent on the pump power after $\sim 70\%$. At 20 bar, the maximum quantum efficiency is 74%, corresponding to 17.6 μJ pulse energy. To the best of our knowledge, both pulse energy and quantum efficiency here are the highest compared to all reported gas-filled ARHCF lasers beyond 3 μm , including both fiber laser and solid-state laser pumping.

4. MULTI-WAVELENGTH NIR RAMAN LASER GENERATION

Besides the presented vibrational Raman laser in last section, the rotational SRS process of H_2 can also be adopted for laser frequency shift. The efficiency of rotational SRS can be significantly enhanced by simply changing the pump polarization from linear to circular state³⁴. Due to its short frequency shift coefficient of 587 cm^{-1} and the wide transmission band of ARHCF, this property is suitable for cascaded Raman laser generation with multiple wavelengths. Here we demonstrate a high pulse energy multi-wavelength Raman laser spanning from 1.53 μm up to 2.4 μm by replacing the above ARHCF with a 5 m long nested ARHCF. The HWP is replaced by a quarter-wave plate before the nested ARHCF to convert the pump light from linear polarization to circular polarization and to optimize the polarization orientation. The

nested ARHCF has a core diameter of 37 μm , surrounded by seven nested capillaries, with wall thicknesses of ~ 406 nm and ~ 630 nm for the outer and inner capillaries, as shown by Figure 3(a). This structure enables a low fiber loss at the NIR region, while preserves a high loss after 3 μm , therefore facilitating the cascaded rotational SRS by preventing the extra pump energy consumption with the vibrational SRS (see Figure 3(b)). At the output side of the nested ARHCF, the power of each individual Stokes line was measured by extracting them with a series of long-pass filters.

Figure 3. Multi-wavelength NIR Raman laser generation in H_2 -filled ARHCF. (a) SEM images of the nested ARHCF. (b) Simulated loss spectrum of the nested ARHCF. (c) Experimental result showing the evolution of the Raman laser spectrum as a function of H_2 pressure. (d) Optical spectrum of the cascaded Raman fiber laser at 21 bar H_2 . (e) Total output power (black) and total Raman power of all Stokes lines (red) versus pressure. (f) Calculated quantum efficiencies of each Raman Stokes line versus pressure.

The evolution of the measured Raman spectrum in terms of pressure was investigated at the maximum pump power and proper orientation of the quarter-wave plate, as presented in Figure 3(c). An optical spectrum analyzer (OSA) (Spectro 320, Instrument Systems) was used to record the spectra. Initially, only the first-order rotational Stokes line at 1683 nm was observed at the pressure of ~ 1 bar. Increasing the gas pressure increases the Raman gain coefficient of the steady-state Raman regime¹⁵ and suppresses the transient Raman regime¹⁶, which has a lower gain coefficient than the steady-state regime. As a result, the intensity of the first-order Stokes line first continuously increases until it is sufficient to serve as a new pump to generate the second-order Stokes line at 1868 nm. Similarly, the third-order Stokes line at 2099 nm was formed from the second-order Stokes line and then increases in intensity and finally leads to the formation of a fourth-order Stokes line at 2394 nm when the pressure increases to ~ 11 bar. It can be seen that the emergence of new Stokes lines is always accompanied by a decrease in the intensity of the previous order Stokes line, indicating that the last order Stokes line plays the role of the pump for the generation of the new Stokes line. Because the fifth-order Stokes line at ~ 2.8 μm could not be formed due to the high fiber loss of 105 dB/m, the intensity of the fourth-order Stokes line continuously increases with pressure up to ~ 21 bar, which is the maximum available pressure of the experimental setup. Figure 3(d) shows the measured spectrum at 21 bar, where the pulse energies are 2.1, 4.3, 7.9, and 8.2 μJ at 1683, 1868, 2099, and 2394 nm, respectively. Figure 3(e) shows the evolution of total power (includes all Stokes lines and residual pump) and total Raman laser power (includes all Stokes lines, but not the pump) as a function of gas pressure. It can be seen that the former continuously drops down as the gas pressure increases, indicating a energy transfer from the pump to the Raman laser Stokes lines. Meanwhile, the latter first quickly increases to a maximum value of 268 mW, but then drops slowly again due to the saturation of the Raman gain coefficient with increase in H_2 pressure¹⁵. This is mainly attributed to the low photon energy at longer wavelengths and the increased fiber propagation loss experienced by the higher-order Stokes lines at longer wavelengths. Figure 3(f) shows the quantum efficiency for each Raman Stokes line. The maximum quantum efficiencies at 1683 nm, 1868 nm, 2099 nm, and 2394 nm are 50.5%, 43.5%, 52.5%, and 38.1% respectively, corresponding to pulse energies of 18.5 μJ , 14.4 μJ , 14.1 μJ and 8.3 μJ , respectively. One can see that the pulse energy of each Stokes line can easily reach the micro-joule level. To the best of our knowledge, this is the first time to achieve micro-joule pulse energy over four cascaded Raman Stokes lines in such a broad spectral range.

5. CONCLUSION

We summarized our recent works on gas-filled ARHCF lasers, including a SC generation spanning from 200 nm to 4 μm , and high energy infrared Raman lasers in single wavelength and multiple wavelengths. These results reveal the huge potential of the gas-filled ARHCF technology for laser emission from DUV to MIR, and provide diverse and powerful solutions for applications such as gas spectroscopy, imaging, etc.

ACKNOWLEDGEMENTS

This work is supported by the Danmarks Frie Forskningsfond Hi-SPEC project (Grant No. 8022-00091B), Multi-BRAIN project funded by the Lundbeck Foundation (R276-2018-869), ECOMETA (Grant No. 6150-00030B) and US ARO (Grant No. W911NF-19-1-0426). Abubakar I. Adamu is now with Lumenisity Ltd, United Kingdom.

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